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LABORATORY OF NUCLEAR RADIATION

Head: Dr. Sakae Shimizu

The Laboratory of Nuclear Radiation headed by Prof. Sakae Shimizu was established in June 1957. The history and an outline of research activities of the Laboratory in the first decade have been reported in *the Bulletin* of our Institute **44**, No. 6, November 1966 on the occasion of the fortieth anniversary of the Institute. For the last decade the Laboratory has been developing year by year and has achieved many research results. The Laboratory has had visiting scientists from the United States and Europe and has sent members of the research staff to the leading research institutes in Europe. During this period the following physicists joined our group: from September 1969 till July 1970 Prof. L. B. Borst, State University of New York at Buffalo, and Prof. C. Kikuchi, University of Michigan; from September 1972 until January 1973 Prof. H. L. Finston, Brooklyn College of the City University of New York; from November 1972 till November 1973 Dr. Imre G. Kádár, Institute of Nuclear Research of the Hungarian Academy of Sciences, Debrecen. From our laboratory, Dr. Y. Nakayama has been studying experimental nuclear physics and heavy ion reactions in Prof. E. Kankeleit's Laboratory in the Technische Hochschule Darmstadt, and at the Gesellschaft für Schwerionenforschung mbH (GSI) in Darmstadt, West Germany, for more than two years. Dr. H. Mazaki studied high pressure and low temperature physics in Laboratoire des Interactions Moléculaires et des Hautes Pressions, headed by Prof. B. Vodar, in Meudon, France, for ten months. Dr. Y. Isozumi is now working with Prof. R. J. Walen's group in Centre de Spectrométrie Nucléaire et Spectrometrie de Masse, in Orsay, France, and Drs. S. Shimizu, T. Mukoyama, Y. Isozumi, T. Kitahara and Mr. R. Katano have attended international conferences and visited some leading research institutes and universities in the United States and Europe to see their current activities and discuss with these research workers problems of interest. Such an opportunity to work with foreign research scientists has been developed in the last decade. It is a sign of the international recognition of our laboratory.

The Radioisotope Research Laboratory, supervised by Prof. S. Shimizu for twenty years since 1952, has been converted to a new independent research center belonging directly to the University. The Radioisotope Research Center was inaugurated in April 1971 and Dr. Shimizu was appointed Director as an additional responsibility. The Laboratory of Nuclear Radiation is now located in the old building of the Center. Our members work in cooperation with the staff of the Center to operate it as a service for many research workers from other institutes of the University.

RESEARCH PROGRAM

Recently we have been concerned mainly with studies on higher order nuclear phenomena involving atomic shell electrons. These studies are related to various fields of nuclear, atomic, and solid state physics. The problems are summarized as follows:

I. Positron Annihilation with *K*-Shell Electrons

When a positron annihilation takes place with an electron there exist two distinct processes; (a) annihilation by a free electron and (b) by a shell electron strongly bound in an atom. In the former case, at least two photons are emitted because of the conservation law of momentum, while in the latter a *single-quantum annihilation* (SQA) can take place. Total cross sections for SQA with *K*-shell electrons in five elements, ${}_{50}$Sn, ${}_{73}$Ta, ${}_{79}$Au, ${}_{82}$Pb, and ${}_{92}$U, have been measured in the positron energy range from 250 to 400 keV. The total cross sections and the *Z*-dependance of this annihilation process were found to be in fairly good agreement with theoretical values calculated by other workers.

As a competitive process of SQA *radiationless* or *zero-quantum annihilation* (ZQA) would be expected. When SQA arises with one of the *K*- or *L*-shell electrons, owing to the presence of other bound electrons in the atom, it is possible for the excess energy liberated to be used to eject another bound electron from the atom, the annihilation thus taking place without radiation. We observed this ZQA process using lead as a target and 300-keV positrons. The cross section found experimentally is $\sigma_{\mathrm{exptl}}=0.8_{-0.3}^{+0.4}\times 10^{-26}\,\mathrm{cm}^2$ as the sum of those for *K-K, K-L, K-M,* and *L-L* pairs of shell electrons in a lead atom for 300-keV positrons. We have also developed a theory and the calculated cross section is $\sigma_{\mathrm{calc}}=0.727\times 10^{-26}\,\mathrm{cm}^2$. Our experimental value is in agreement with the calculated result within the experimental error. Our work has established this mode of positron annihilation, and has increased our understanding of the process.

In the process of positron annihilation with *K*-shell electrons another possibility may exist that the excess energy liberated would be used to excite the nucleus of the atom involved. We have established experimental evidence for this mode of positron annihilation by *K*-shell electrons, *nuclear excitation by positron annihilalion,* with ^{115}In and 300-keV incident positrons. The experimental cross section observed in this case is of the order of $10^{-25}\,\mathrm{cm}^2$. Further study of this process is now in progress.

Another mode of annihilation of positrons in flight with *K*-shell electrons is expected. We attempted to observe this *two-quantum annihilation* (K-TQA). Using a silver target and a 300-keV positron beam, we have obtained the experimental result $(7.7\pm 6.4)\times 10^{-27}\,\mathrm{cm}^2/\mathrm{sr}^2$ for the double-differential angular cross section at 30° and $-100°$ for each annihilation photon with respect to the incident positron direction.

Further experimental work on the inner-shell ionization by positron impact is in progress, using monoenergetic positron beams produced by the double-focusing β-ray spectrometer mounted by a β^+ source.

II. External Effects on the Decay Constant of ^{99m}Tc

In 1964–1966 we worked to observe the effect of chemical bonding on the decay constant of ^{235m}U. Since then, we have continued experimental work in this field.

We have studied the change in the decay rate of the 2.17-keV E3 isomeric transition of ^{99m}Tc ($T_{1/2}$=6.04 h) under the effects of (a) high pressure, (b) the intense internal electric field in the ferroelectric state, and (c) superconducting state of thin samples. In these studies we adopted a differential method to detect the minute changes in the decay constant of the ^{99m}Tc samples.

Ferroelectric samples of $BaTi(^{99m}Tc)O_3$ were prepared, in which some of the Ti atoms in $BaTiO_3$ crystals are replaced by ^{99m}Tc atoms. The decay constants of ^{99m}Tc in samples in the ferroelectric (room temperature) and paraelectric (170°C) states were compared by means of the differential method. The result obtained is: $(\lambda_p-\lambda_f)/\lambda_f=(2.6\pm0.4)\times10^{-3}$, where λ_f is the decay constant for $BaTi(^{99m}Tc)O_3$ in the ferroelectric state and λ_p is that for $BaTi(^{99m}Tc)O_3$ in the paraelectric state.

An experimental investigation of the influence of high hydrostatic pressure on the decay constant of ^{99m}Tc had been performed. By the use of a multianvil high-pressure apparatus constructed by ourselves, a carrier-free source of ^{99m}Tc was compressed at a pressure of 100 kbar. The observed decay rate of the compressed ^{99m}Tc source was compared with that of a standard ^{99m}Tc source in the normal uncompressed state. The fractional increase in the decay constant is $\Delta\lambda/\lambda=(4.6\pm2.3)\times10^{-4}$.

We have been studying the change in the decay constant of ^{99m}Tc under the influence of superconducting state, using a specially designed double ionization chamber for the 140-keV γ rays. We have recently found the relative change in the decay constant of this isomer at room temperature and below the critical temperature (7.6 K) to be zero within experimental error. This result, which does not agree with that reported in the United States, raises question concerning the influence of the superconducting state on the 2.17-keV E3 transition of ^{99m}Tc. Fuller accounts of this work will be published in the near future.

III. *K*-Shell Internal Ionization Accompaying Nuclear Decay

Internal ionization in the K-shell during nuclear $\boldsymbol{\beta}$ *decay* was studied experimentally. The K x rays produced were measured in coincidence with emitted electrons for various segments of the β spectra of ^{147}Pm and ^{63}Ni, using 4π detection geometry for these electrons; the sources were mounted in the electron detectors, a split anthracene scintillation crystal and a gas-flow proportional counter, respectively. The energy-dependent ionization probability was measured as a function of $E_\beta{}^\circ$, which is defined as a sum of energies of the β particle and emitted K electron plus the K-shell binding energy of daughter atom. Then, we proposed a new type of experiment to obtain directly the sum of the energies of electrons (β particles plus emitted K-shell electrons) in coincidence with emitted K x rays in the β decay of ^{63}Ni using two proportional counters.

We have also developed an improved theoretical treatment of the phenomenon by correcting a mistake in the theory of Law and Campbell. Using the improved formulation and adding simple corrections for the K-shakeup contribution and for the correlation effect between K electron and other bound electrons, the total K-hole creation probability per β decay has been evaluated for 24 interesting nuclides. There exist distinct disagreements between our calculated values and recent experimental data;

the measured probabilities are systematically larger than our calculated ones for medium- and high-Z nuclides. This clearly indicates that the theory of K-shakeoff, as here corrected, is not sufficient to explain the recent experimental data. This is contrary to the conclusion recently presented by many other workers that the K-shakeoff plus -shakeup process is the predominant mechanism of K-shell internal ionization and excitation in β decay. Essential points of our theory have already been published in three articles in *Lettere al Nuovo Cimento*. Detailed discussion on this problem will be published in the near future.

The K-shell internal ionization in K capture was studied experimentally for ^{55}Fe; the energy spectrum of K electrons ejected from ^{55}Fe during K-capture decay has been measured in the energy range from 5 to 180 keV. In addition, a value of the total K-shell internal ionization probability per K capture has been estimated experimentally. The phenomenon was observed by the triple-coincidence (K-x—ejected e^-—K-x) experiment using a gas proportional counter for ejected electrons and two gas proportional counters for Mn K x rays. For the higher-energy region of ejected electrons, the confinement of the electrons within the sensitive volume of the electron counter was achieved by applying a magnetic field. The L-shell contributions were rejected completely by the triple-coincidence technique.

Observations of the double K-hole creation in the internal conversion, *i.e., K-shell internal ionization and excitation in the K-internal conversion process,* were recently performed with ^{141}Pr and ^{137m}Ba by means of the coincidence technique using two high-resolution Si(Li) detectors.

On the other hand, we studied theoretically the K-shell internal ionization and excitation during K-capture and K-conversion processes; the phenomena were treated relativistically by the use of screened relativistic hydrogenic wave functions. The calculated values we obtained were compared with our experimental results and found to be in fairly good agreement. Our theoretical studies have recently been published in a series of papers.

IV. Mössbauer Effect Studies

A Mössbauer spectrometer with a large driving force was designed and constructed, by which a sample could be supported in a liquid-helium cryostat, and Mössbauer studies were carried out on a ferromagnetic Ni-Cu alloy near the transition temperature and on SnI_4.

A new Mössbauer detector assembly, consisting of two thin proportional counters combined together, was designed. One is used to detect Fe K x rays emitted after the K-shell internal conversion process of the resonantly excited 14.4-keV level of ^{57}Fe, while the other is used to detect electrons emitted by the conversion process and by the Auger effect after the process. Using a special type of thin proportional counter with pure helium gas to detect scattered electrons, we have recently succeeded in observing the spectra of emitted electrons accompanying the Mössbauer effect on samples cooled to liquid nitrogen temperature. Specially designed electron proportional counters are also now being used to study the Mössbauer effect of the uppermost surface of some compounds at several hundred degrees.

V. Experimental Nuclear Physics

In this period only two experimental studies were performed. One of the nuclear Raman scattering lines from ^{181}Ta using Li(p, γ) γ rays was observed in 1967. This was an experiment in the early state of study on this phenomenon.

The other work, performed for about four years from 1967 to 1971, was the experiment to measure the probability and the energy distribution for the two-photon decay from the 1.76-MeV 0^+ first excited state to the 0^+ ground state of ^{90}Zr. A sophisticated technique was used; a sum-coincidence technique with an antidetector and large lead shields. By this formidable work an experimental value of the ratio of two-photon decay to the sum of internal-pair decay and internal-conversion decay, $T_{\gamma\gamma}/T_{\pi+e}$, was obtained as $(5.1 \pm 2.5) \times 10^{-4}$. Furthermore, from the observed spectrum of one of two photons, it was suggested that two-photon decay takes place as an (El, El) transition via giant-dipole-resonance states.

VI. Gamma-Ray Irradiation Facility

In 1969 a new compact 2000 Ci cobalt-60 irradiation facility was designed and installed. Studies on effects of γ-ray irradiation in many fields, using this facility, have been published every year in the special issue of our Bulletin entitled "*Physical, Chemical, and Biological Effects of Gamma Radiation*".

VII. Low Temperature Physics and High Speed Rotation

In addition to the study fields mentioned above, we have recently been working with some experiments in low temperature physics and to develope a technique to get ultra-high centrifugal fields by high speed rotation of steel balls.

To achieve very low temperature below 100 mK, we have prepared a device including a superconducting magnet. With this device, properties of superconducting materials as affected by magnetic impurities will be investigated. The study of superconductivity asking radioactive materials is expected to give new insight into its nature and application.

An apparatus to produce ultra-high centrifugal fields higher than 10^8 g has been developed by applying the magnetic levitation method to small steel balls in high vacuum which are rotated by the rotating magnetic field. We have recently achieved 15.85×10^4 rps with a small steel ball 2.0 mm in diameter. This corresponds to a centrifugal field of 1.00×10^8 g at the equator of the ball. We are now endeavouring to get higher fields by modifying the electronics of the device. We plan to apply this technique to search for the effect of high centrifugal fields on the decay of some radioactive nuclides as well as to studies in solid state physics and others.

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